Effect of quantum confinement on exciton–phonon interactions

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We investigate the homogeneous linewidth of localized type–I excitons in type–II GaAs/AlAs superlattices. These localizing centers represent the intermediate case between quasi–two–dimensional (Q2D) and quasi–zero–dimensional localizations. The temperature dependence of the homogeneous linewidth is obtained with high precision from microphotoluminescence spectra. We confirm the reduced interaction of the excitons with their environment with decreasing dimensionality except for the coupling to LO phonons. The low–temperature limit for the linewidth of these localized excitons is five times smaller than that of Q2D excitons. The coefficient of exciton–acoustic phonon interaction is $5 \sim 6$ times smaller than that of Q2D excitons. An enhancement of the average exciton–LO phonon interaction by localization is found in our sample. But this interaction is very sensitive to the detailed structure of the localizing centers.

The homogeneous linewidth of exciton luminescence is one of the most important features in excitonic dynamics in semiconductors, since it contains directly the information about the interactions between excitons and their environment. During the past two decades, the homogeneous linewidth of excitons in several kinds of quantum well and superlattice systems has been investigated extensively in both time and frequency domains. In the time domain, the excitonic dephasing time was measured from four–wave mixing (FWM), and then the homogeneous linewidth could be deduced.[1, 2, 3, 4, 5] In the frequency domain, the linewidth was measured directly from photoluminescence,[6, 7, 8, 9] transmission, reflection or absorption[10, 11] and Raman spectroscopy[12]. By modeling of experimental data, extensive information about interactions between excitons and acoustic phonons, LO phonons, free carriers and other excitons has been deduced. In these investigations, excitons are quasi–two–dimensional (Q2D). That is, they can move freely in the wells or are localized weakly with a localization energy of several meV. On the other side, the homogeneous linewidth of quasi–zero–dimensional (Q0D) excitons confined in quantum dots, with localization energy of several hundreds of meV, has been studied by spatially resolved measurements.[13, 14] The comparison of these two kinds of excitons provides information about the influence of quantum confinement on the interactions between excitons and their environment.

In this paper, we report investigations on homogeneous linewidth of single type–I localized excitons in GaAs/AlAs superlattices which have a global band alignment of type II. The localization energies of these centers are several tens of meV. Thus we can regard these localized excitons as intermediate in dimensionality between Q2D excitons and Q0D excitons. Furthermore, since the investigated centers are found in a small area (1 μm in diameter) of the same sample, we can rule out any artificial effects which come about when comparing different samples. This enables us to discuss the influence of localization on exciton-phonon interactions by comparing these centers, without disturbed by other artificial effects.

The localized excitons are studied by microphotoluminescence (μ–PL). The spectral and spatial resolutions are sufficient to detect luminescence from individual localizing centers. Our experimental setup consists of a He flow cryostat with the sample mounted close to a thin window. This allows the use of a microscope objective to image (magnification 20×) the excited spot on the sample onto a pinhole. The pinhole defines the spatial resolution. We use a 20–μm pinhole in the present experiments, which corresponds to 1–μm detected area on the sample's surface. The pinhole is imaged onto the entrance slit of a 0.75–m focal length double grating spectrometer. We use a cooled CCD to record the spectra with a spectral resolution of 30 μeV. The sample is nonresonantly and globally excited by a He–Ne laser. The excitation intensity is about 1 W/cm^2 for all of the temperature–dependent measurements. During the measurement, the temperature of the sample is measured with a diode temperature sensor in good thermal contact. The temperature is stabilized by the He flow and heating to a fluctuation of less than 0.2 K. The measurements are performed in the range of $7 \sim 80$ K. We study two samples: (i)140 periods of GaAs(3 nm)/AlAs(2.8 nm) and (ii)140 periods of GaAs(2.3 nm)/AlAs(2.3 nm). Both samples have a type–II band alignment, i.e., the unperturbed conduction-band minimum is in the AlAs layer and the valence–band maximum is in the GaAs layer. Details about the growth and the interface properties of the samples have been reported previously.[15] The two samples yield quite similar results concerning the exciton–phonon coupling. Thus we will only present data of sample (i).

Figure 1 reviews the luminescence properties of the sample at 20 K. The spatially integrated PL spectrum [Fig. 1(a)] is composed of a zero–phonon line at about 1.782 eV and phonon sidebands at the low–energy side. Luminescence intensity maps [Fig. 1(b)] show an inhomogeneous distribution of the emission intensity. We can find bright spots of about 1 μm in diameter. This size corresponds to the resolution of the objective in our μ–PL system. The actual size of the bright spots was determined to be about $250 \sim 300$ nm [full width at half maximum (FWHM)] by scanning near–field optical microscopy with resolution of 100 nm.

Figure 1(c) shows the μ–PL spectrum of one of the bright spots. Different from Fig. 1(a), the spectrum

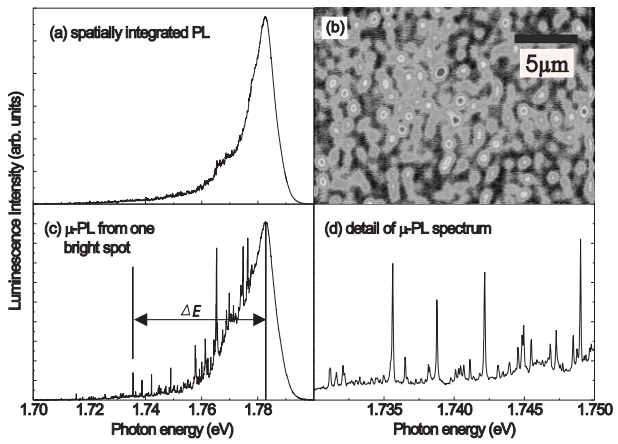

FIG. 1: Luminescence of GaAs/AlAs superlattices at 20 K under excitation of He–Ne laser with excitation intensity of about 1 W/cm^2. (a) Spatially integrated PL spectrum; (b) Intensity maps; (c) The μ–PL spectrum of one of the bright spots; (d) Details of (c). The intensity maps are recorded by blocking the scattered light from the laser, and spectrally integrating the luminescence from 1.7 to 1.8 eV.

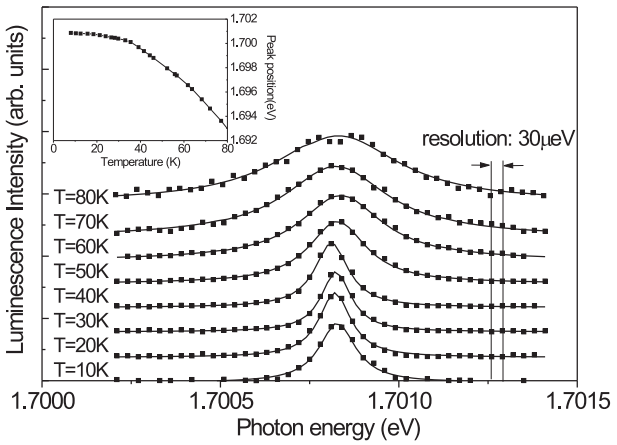

FIG. 2: An example of the measured narrow lines (squares) and the corresponding Lorentzian fit (solid line). The peak positions have been shifted for better illumination. For the actual positions under different temperatures, see the inset.

is dominated by local emission from the bright spot. On the smooth background, spectrally narrow lines are superimposed.[16] Figure 1(d) provides a closer sight of these narrow lines. We have found that the narrow lines observed in Fig. 1(c) could be divided into two groups according to their different temperature behaviors.[16] For the lines on the low–energy side of the zero–phonon line, i.e., 1.75 $\sim$ 1.782 eV, the spectral weight shifts red with rising temperature, and their integrated intensity drops. These lines stem from localized type–II states. For the lines in the spectral range of the AlAs LO–phonon replica, i.e., below 1.74 eV, the spectral weight does not change significantly, and their intensities increase exponentially with temperature up to 50 K. We have proved that, although the global band alignment of this sample is type II, the layer thickness fluctuations give rise to local changes in the band alignment toward type I. Recombination of excitons localized in these type–I centers is the origin of the narrow lines in the energy range of 1.69 $\sim$ 1.74 eV. The population mechanism of these localized states has been proved to be electron tunneling from AlAs layers to GaAs layers.[16] Since some of these narrow lines are well separated in energy, we can resolve each of them without serious disturbance by adjacent lines. Thus we can analyze the luminescence from single localizing centers in the GaAs layers.

In order to investigate the temperature dependence of the linewidth of excitons localized in the type–I centers, μ–PL spectra from several bright spots were measured in the temperature range of 7 $\sim$ 80 K. To check the possible spectral wandering during the integration, we measured the spectra with different integration times. The spectral shape and the linewidth keep unchanged as we vary the integration time in the range of 50 ms $\sim$ 30 s. We also measured the sequence of the spectra with an integration time of 50 ms and an interruption time of 1 s. We didn't find any change of the peak position among these spectra. Thus, the spectral wandering of the sample can be neglected. The narrow lines were fitted by Lorentzian line shapes to obtain the linewidth (FWHM) in each temperature. An example of the narrow lines and the fitting curves is shown in Fig. 2. Recently, Besombes *et al.*[17] found that the line shape of the luminescence from strongly confined CdTe quantum dot deviates from Lorentzian shape with increasing the temperature. The whole spectrum is composed of a zero–phonon line and an additional acoustic–phonon sideband which results from lattice relaxation due to exciton-phonon coupled states. In InAs/GaAs system, both Lorentzian[18] and the non-Lorentzian[19] line shapes have been observed recently. For GaAs quantum dots, the PL spectrum has been shown to be of Lorentzian line shape.[20] Also in our experiments on centers of intermediate confinement in GaAs/AlAs superlattices, we find no indications of additional phonon sidebands. The spectral line shape can be well fitted by Lorentzian function up to 80 K (see Fig. 2).

In general, the temperature dependent homogeneous linewidth of the exciton resonance is written as (see, e.g., Ref. 21)

$$\Gamma_{homo}(T) = \Gamma_0 + \gamma_{AC}T + \gamma_{LO}[\exp(\hbar\omega_{LO}/k_BT) - 1]^{-1} \quad (1)$$

where the term linear in temperature is due to exciton scattering with acoustic phonons, and the term nonlinear in temperature is due to interactions with LO phonons. The coefficients γ_{AC} and γ_{LO} represent the strength of the exciton–acoustic–phonon interaction and exciton–

LO–phonon interaction, respectively. The first term in Eq. (1) is the low–temperature limit of the linewidth. The temperature dependence of the linewidth deduced from the Lorentzian fits was then fitted by Eq. (1), for several well–separated narrow lines. We show one of the fitting results in Fig. 3, as an example. The contributions to the linewidth from acoustic–phonon scattering and LO–phonon scattering are also shown in this figure (short–dashed and dotted lines, respectively). The dashed line represents low–temperature limit of the linewidth. For this narrow line, the fitting parameters are Γ_0 = 42 μeV, γ_{AC} = 1.2 μeV/K, and γ_{LO}= 85.7 meV. Since the fitting contains three free parameters, it is necessary to check the sensitivity of these parameters on fitting process. For this purpose, we vary each of these parameters from its optimal value, to check the variation of mean–square deviation between fitting curve and experimental data. We find that a 10 % deviation of Γ_0, γ_{AC}, or γ_{LO} from their optimal values corresponds increase of 60 %, 94 % or 580 % in the mean–square deviation, respectively. This result insures the safety of the fitting process. Another possible problem in extracting the parameters from the fitting is whether the temperature range (7 $\sim$ 80 K) is large enough for an accurate determination of the parameters, especially for the γ_{LO}. In the investigations of the Q2D excitons, the linewidths were measured up to 150 K (Ref. 6) or even room temperature[4, 9, 10]. In the present study, however, the luminescence of these localized excitons at a temperature above 80 K is too weak for an accurate determination of the linewidth. To check the possible errors introduced by this relatively small temperature range, we redraw the results obtained by the larger temperature range measurements,[4, 6, 9, 10] and fit by Eq. (1) all the data in the whole temperature range or only the data obtained below 80 K, respectively. We find that for all of the results checked, the difference of the parameter values obtained from the two fits using different ranges of data is less than 10 % for γ_{LO} and less than 3 % for γ_{AC}. This proves the temperature range of 7 $\sim$ 80 K is enough for the determination of the linewidth–temperature curve with a satisfactory accuracy. Furthermore, the data density in the present study (about 50 data points in the temperature range of 7 $\sim$ 80 K) is high enough for an accurate fitting.

In Figure 4(a), we list the fitting results of Γ_0, γ_{AC}, and γ_{LO} of the narrow lines analyzed in the present study. In order to distinguish the narrow lines, we define ΔE as the energy difference between the corresponding narrow line and the peak of zero–phonon line. We show an example of this definition for one narrow line in Fig. 1(c). Such a quantity is temperature independent. We note that ΔE is close to, but not exactly, the localization energy of the corresponding center, since we cannot regard the zero–phonon–line as the mobility edge exactly. However, such a difference will not influence our discussions. For comparison, we list in Fig. 4(b) the available data of GaAs Q2D excitons deduced from FWM, photoluminescence,

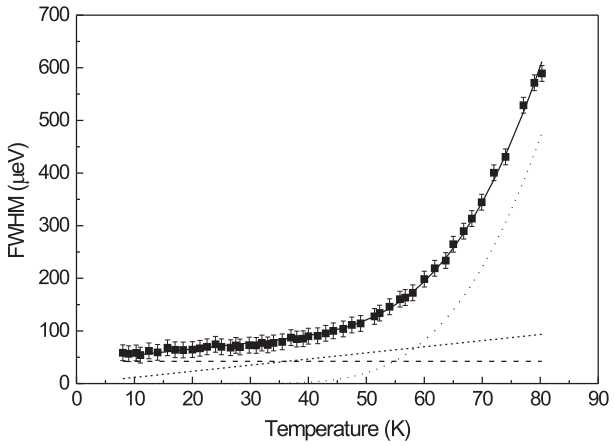

FIG. 3: Temperature dependence of the homogeneous linewidth of one narrow line. The experimental data (squares) were fitted by Eq. (1) (solid line). The contributions to the linewidth from acoustic–phonon scattering and LO–phonon scattering are also shown (short–dashed and dotted lines, respectively). The dashed line represents the low–temperature limit of the linewidth.

or other methods, as a function of the thickness of the GaAs layers. The results of GaAs bulk and superlattices are also listed in this figure, but not included in the calculations of average values, which are shown as dashed lines in Fig. 4(b). In the viewpoint of quantum confinement, we regard the localized excitons investigated here as the intermediate case between Q2D excitons in quantum wells and Q0D excitons in quantum dots. In the following, we will compare the Γ_0, γ_{AC}, and γ_{LO} of localized excitons obtained in the present study with that of the other two cases, to discuss the influence of confinement on exciton–phonon interactions in semiconductors.

At first, we discuss the low–temperature limit of the linewidth. We obtain the average value of Γ_0 to be 0.057 ($\pm$0.014) meV for these localized excitons [dashed line in Fig. 4(a)I]. This value is five times smaller than the average value of Q2D excitons [dashed line in Fig. 4(b)I]. That indicates that the additional in–plane confinement in localizing centers reduces Γ_0. In order to investigate the contribution of intercarrier scattering to this linewidth, we measured the excitation intensity dependence of the linewidth at 7 K. In the intensity range of 1 $\sim$ 10 W/cm^2, the linewidth keeps unchanged, while in the range of 10 $\sim$ 5000 W/cm^2 the linewidth increases slowly with a slope of 0.01 $\sim$ 0.02 μeV/(W/cm^2). Due to the complicated population mechanism of these localizing centers (electron tunneling from AlAs layers to GaAs layers), we are not able to relate the excitation intensity to the actual carrier density in the sample. However, we can conclude that the excitation intensity used in the temperature–dependent measurement is quite low, and the intercarrier interaction can be neglected. The in-

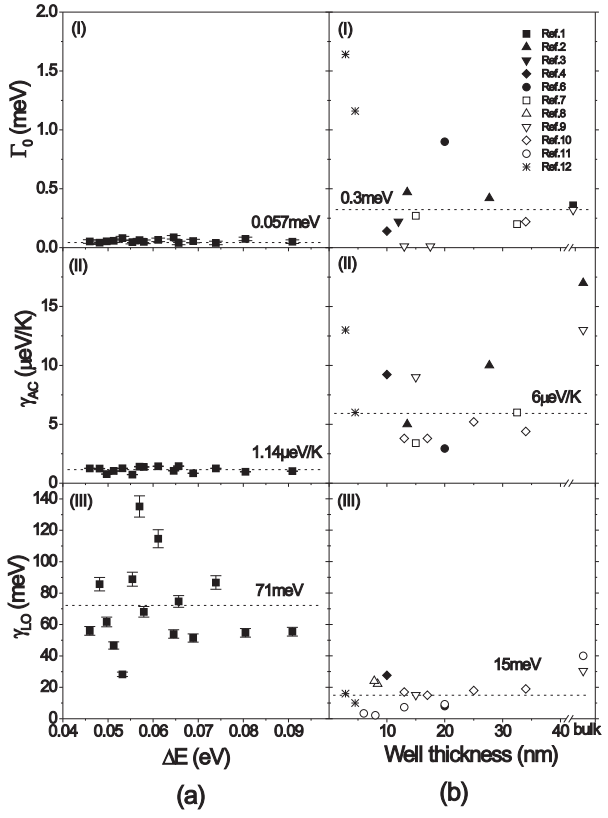

FIG. 4: (a) Low–temperature limits of linewidth (I), exciton–acoustic–phonon interaction coefficients (II) and exciton–LO–phonon interaction coefficients (III) gained by fitting the measured $\Gamma \sim T$ relations (see Fig. 3). The error bars show the uncertainty of the fitting due to a limited temperature range, as discussed in the text. The dashed lines represent the average values; (b) The corresponding available values for quasi–two–dimensional excitons in GaAs quantum wells and excitons in GaAs bulk and superlattices. References for all data points are listed in (b)I.

trinsic lifetimes of excitons in quantum wires and quantum dots have been calculated to be several hundreds of picosecounds.[22] However, recent experiment reveals a 16–ps intrinsic lifetime of excitons in GaAs quantum dots.[23] The linewidth obtained in the present study corresponds to a lifetime of 22 ps, which is consistent with this new finding.

Second, we discuss the influence of confinement on exciton–acoustic–phonon interaction. It has been found before[2, 12] and confirmed again recently[9] that γ_{AC} of Q2D excitons is smaller than that in bulk GaAs [Fig. 4(b)II] . The average value of γ_{AC} of the localized excitons obtained in the present investigation is 1.14 ($\pm$0.24) μeV/K, about 5 $\sim$ 6 times smaller than that of Q2D excitons [dashed lines in Figs. 4(a)II and 4(b)II]. The result suggests a reduction of exciton–acoustic–phonon interaction by localization. In quantum wires, such a reduction has been found by direct comparison of free and localized excitons in FWM measurements.[24] Furthermore, in quantum dots, the homogeneous linewidth has been found to be almost constant up to 50 K.[13, 14] These results suggest the extremely small γ_{AC} for Q0D excitons. The weaker acoustic–phonon interaction with Q0D excitons than with Q2D excitons has also been confirmed in II–VI systems (see, for example, Ref. 17). The whole evolution discussed above, from bulk via Q2D excitons to localized excitons (this study) and to Q0D excitons, implies strongly that the interaction between exciton and acoustic phonon is steadily reduced by increasing confinement. Such a behavior is consistent with previous theoretical predictions.[25, 26] In a confined system, the final state of phonon scattering is not always available due to the discrete energy level scheme. Thus, by increasing the quantum confinement, the appearance of the discrete energy levels induces a decrease of the acoustic phonon interaction. We note that when the confinement is so strong that the energy level space is larger then the thermal energy k_BT, a further increase of the confinement does not further reduce the interaction, since the level space has already been large enough for this bottleneck effect. In this regime, additional effects like lattice relaxation[17] can influence the dependence of the acoustic phonon interaction on the quantum dot size. Theoretical calculations revealed an increase, rather than decrease, of the acoustic–phonon coupling when further reducing the size of the quantum dots in this regime.[17, 27]

The parameters Γ_0 and γ_{AC} of the localized excitons obtained here are almost independent of ΔE in the range of 0.04 $\sim$ 0.09 eV [Figs. 4(a)I and 4(a)II]. But, for γ_{LO}, we find a totally different behavior in the same energy range. The values of γ_{LO} vary in the range of 30 $\sim$ 140 meV, with no obvious systematic dependence on ΔE. The fluctuations of Γ_0 and γ_{AC}, which are also obtained in the same fitting process, are all less than 25 %. We attribute the observed scattering in the homogeneous linewidth to an intrinsic feature of the exciton–LO–phonon coupling. In localizing centers, the energy level scheme of excitons is determined by the detailed structure of the center. Due to the monochromatic feature of the LO–phonon dispersion, the exciton–LO–phonon scattering rate depends sensitively on the level scheme. For the center in which the energy level scheme matches the LO–phonon energy well, a strong coupling is observed. In contrast to the LO phonons, the dispersion of the acoustic phonons distributes over a relative wide energy range. Thus, the exciton–acoustic–phonon scattering rate is less sensitive to the detailed structure of the localizing centers. In fact, we do not find the pronounced resonant behavior for the acoustic–phonon coupling [see Fig. 4(a)II].

In the strongly confined quantum dots, the explicit size and shape of the localizing potential determines the spatial extension and anisotropy of the electron–hole wave function as well as the electron–hole over-

lap. This has significant influence on the exciton–phonon interaction.[28, 29] In strongly confined CdTe quantum dots, a mixing of the exciton and acoustic–phonon modes, which cannot be described by perturbation treatment, has been proposed.[17] That is, the exciton locally distorts the lattice of the dot. This lattice distortion is important for small quantum dots which sizes are comparable with the exciton Bohr radius. For example in II–VI and InAs/GaAs systems, an induced non-Lorentzian broadening have been observed.[17, 19] However, the localizing centers studied here are much larger than the Bohr radius. Thus the distortion is less important, and we do not observe strong deviations from a Lorentzian lineshape even at a temperature of 80 K. For the same reason, the influence of the potential size and shape on the electron–hole wave function is also less pronounced than that in strongly confined quantum dots. So we observe only small variations in the acoustic–phonon coupling strength among these localizing centers with different sizes and shapes.

Despite of the scattering behavior, we can still deduce the enhancement of the exciton–LO–phonon interaction in localized excitons with respect to Q2D excitons. The average value of γ_{LO} is 71 meV, about five times larger than that of Q2D excitons [Figs. 4(a)III and 4(b)III]. The enhancement of exciton–LO–phonon interaction by localization induced by alloying fluctuations in alloy $GaAs_{1-x}P_x$ has been found by resonant Raman spectroscopy.[30] A similar enhancement was also found in GaN quantum wells.[31] In those investigations, the LO–phonon replica was used to detect the exciton–phonon interaction. In the present study, we detected luminescence from excitons localized by thickness fluctuations by μ-PL. The agreements between different experimental methods as well as the different origins of localization confirm that the additional in–plane confinement on excitons enhances the exciton–LO–phonon interaction.

Up to now, the exciton–LO phonon interaction in quantum dots is still an open problem. In CdTe quantum dots, Besombes *et al.*[17] found that the exciton–LO–phonon scattering is not efficient up to 60 K, while Heitz and co–workers[28, 32] observed enhanced exciton–LO–phonon interaction in InAs/GaAs self–organized quantum dots by measuring the phonon–assisted exciton transitions. The Huang–Rhys parameter was found to be five times larger than in bulk InAs. The enhancement was attributed to the quantum confinement and piezoelectric effect. Our result confirms qualitatively the latter finding. According to the extremely sensitive dependence of exciton–LO–phonon interaction on the detailed structures of localizing centers, we suggest that much care should be taken when comparing experimental results of this interaction in different quantum dot samples, since the detailed structure of the dots can be totally different, and this may influence the strength of the interaction to a great extent.

In summary, we have measured the homogeneous linewidth of type–I localized excitons in type–II GaAs/AlAs superlattices using μ–PL. These excitons, with a localization energy of several tens of meV, can be regarded as intermediate case between Q2D excitons (free or weakly localized excitons in quantum wells) and Q0D excitons in quantum dots with confinement energy of several hundred meV. The low–temperature limit of the linewidth, Γ_0, of these localized excitons is found to be five times smaller than that of Q2D excitons. We obtain a $5 \sim 6$ times smaller exciton–acoustic–phonon interaction coefficient, γ_{AC}, for the localized excitons with respect to that of Q2D excitons. Together with a comparison of exciton data in bulk and quantum dots, the reduction of exciton–acoustic–phonon interaction by confinement is confirmed. In contrast to the results on Γ_0 and γ_{AC}, which are independent of localization energy, the coupling to LO phonons, γ_{LO}, shows strong variations. This finding is attributed to the strong influence of the energy level scheme on the exciton–LO–phonon coupling. In average, we confirm an enhancement of exciton–LO–phonon interaction by localization.

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